

WHAT IS CLAIMED IS:

5 ①. A process for producing an electrode material for a rechargeable lithium battery in which oxidation-reduction reaction is used, said process comprising the steps of:

10 (1) mixing at least one kind of a metal compound (a) selected from a group consisting of metal salts and metal complexes of a metal (a') capable of being electrochemically alloyed with lithium, at least one kind of a transition metal compound (b) selected from a group consisting of transition metal salts and transition metal complexes of a transition metal (b') and a complexing agent (c) with a solvent (d) to obtain a mixed solution,

15 (2) mixing a reducing agent (e) with said mixed solution to obtain a mixture, and

20 (3) oxidizing said reducing agent in said mixture to reduce ion of said metal (a') and ion of said transition metal (b') to obtain an amorphous alloy material capable of being electrochemically alloyed with lithium as said electrode material.

2. The process according to claim 1, wherein said amorphous alloy material is a powdery amorphous alloy material containing an amorphous metallic material.

25 3. The process according to claim 2, wherein said

powdery amorphous alloy material has a main peak having a half width of more than 0.2° in X-ray diffraction using $K\alpha$ -rays of Cu as a radiation source.

4. The process according to claim 2, wherein said
5 powdery amorphous alloy material has a main peak having a half width of more than 0.5° in X-ray diffraction using $K\alpha$ -rays of Cu as a radiation source.

5. The process according to any of claims 2 to
4, wherein said powdery amorphous alloy material has a
10 peak appeared in a range of $2\theta = 25^\circ$ to 50° in X-ray diffraction using $K\alpha$ -rays of Cu as a radiation source, having a half width of more than 0.2° .

6. The process according to any of claims 2 to
4, wherein said powdery amorphous alloy material has a
15 peak appeared in a range of $2\theta = 25^\circ$ to 50° in X-ray diffraction using $K\alpha$ -rays of Cu as a radiation source, having a half width of more than 0.5° .

7. The process according to claim 2, wherein said
powdery amorphous alloy material has a crystallite size
20 calculated from X-ray diffraction analysis, which is less than 50 nm.

8. The process according to claim 2, wherein said
powdery amorphous alloy material has a crystallite size
calculated from X-ray diffraction analysis, which is
25 less than 20 nm.

9. The process according to claim 1 or 2, wherein said metal (a') comprises at least one kind of a metal selected from a group consisting of Bi, In, Pb, Si, Ag, Sr, Ge, Zn, Sn, Cd, Sb, Tl, and Hg.

5 10. The process according to claim 1 or 2, wherein said metal (a') comprises at least one kind of a metal selected from a group consisting of Bi, In, Pb, Zn, Sn, Sb, and Tl.

10 11. The process according to claim 1 or 2, wherein said metal (a') substantially comprises Sn.

15 12. The process according to claim 11, wherein said amorphous alloy material contains an amorphous Sn·A·X alloy with a substantially non-stoichiometric ratio composition, with A being at least one kind of a transition metal element, and X being at least one kind of an element selected from a group consisting of B, C, N, O, P, and S, where the element X is not always necessary to be contained, and said amorphous Sn·A·X alloy has a relationship of $\text{Sn}/(\text{Sn} + \text{A} + \text{X}) = 20$ to 80 atomic% in terms of the atom number of each element.

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25 13. The process according to any of claims 1 to 12, wherein said transition metal (b') comprises at least one kind of a transition metal selected from a group consisting of Cr, Mn, Fe, Co, Ni, Cu, Mo, Tc, Ru, Rh, Pd, Ag, Os, Ir, Pt, Au, Ti, V, Y, Sc, Zr, Nb, Hf, Ta, and W.

14. The process according to any of claims 1 to 12, wherein said transition metal (b') comprises at least one kind of a transition metal selected from a group consisting of Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Ag, Os, Ir, Pt, and Au.

15. The process according to any of claims 1 to 12, wherein said transition metal (b') comprises at least one kind of a transition metal selected from a group consisting of Cr, Mn, Fe, Co, Ni, and Cu.

16. The process according to claim 12, wherein said amorphous alloy material has a peak appeared in a range of $2\theta = 25^\circ$ to 50° in X-ray diffraction using K α -rays of Cu as a radiation source, having a half width of more than 0.2° .

17. The process according to claim 12, wherein said amorphous alloy material has a peak appeared in a range of $2\theta = 25^\circ$ to 50° in X-ray diffraction using K α -rays of Cu as a radiation source, having a half width of more than 0.5° .

18. The process according to claim 12, wherein said amorphous alloy material has a crystallite size calculated from X-ray diffraction analysis, which is less than 50 nm.

19. The process according to claim 12, wherein said amorphous alloy material has a crystallite size

calculated from X-ray diffraction analysis, which is less than 20 nm.

20. The process according to any of claims 1 to 19, wherein said amorphous alloy material has an average particle size in a range of from 0.1 to 2 μm .

21. The process according to any of claims 1 to 19, wherein said amorphous alloy material has an average particle size in a range of from 0.1 to 1 μm .

22. The process according to claim 20 or 21, wherein said amorphous alloy material has a particle size distribution in a range of from 0.01 to 20 μm .

23. The process according to claim 20 or 21, wherein said amorphous alloy material has a particle size distribution in a range of from 0.05 to 1 μm .

24. The process according to claim 20 or 21, wherein said amorphous alloy material has an average value of [longest length of particle]/[shortest length of particle], which is in a range of from 1.0 to 2.0.

25. The process according to claim 20 or 21, wherein said amorphous alloy material has an average value of [longest length of particle]/[shortest length of particle], which is in a range of from 1.0 to 1.5.

26. The process according to any of claims 1 to 19, wherein said amorphous alloy material has a specific surface area of more than 10 m^2/g .

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27. The process according to any of claims 1 to 19, wherein said amorphous alloy material has a specific surface area of more than 30 m²/g.

28. The process according to any of claims 1 to 5 19, wherein said metal compound (a) is soluble in said solvent (d).

29. The process according to any of claims 1 to 27, wherein said metal compound (a) comprises at least one kind of a metal salt selected from a group consisting of 10 chlorides, sulfates and nitrates of said metal (a').

30. The process according to any of claims 11 to 27, wherein said metal compound (a) comprises at least one kind of a metal salt selected from a group consisting of chlorides, sulfates and nitrates of Sn.

31. The process according to any of claims 1 to 15 27, wherein said transition metal compound (b) is soluble in said solvent (d).

32. The process according to any of claims 1 to 27, wherein said transition metal compound (b) comprises 20 at least one kind of a metal salt selected from a group consisting of chlorides, sulfates and nitrates of said transition metal (b').

33. The process according to any of claims 1 to 27, wherein said complexing agent (c) comprises at least 25 one kind of a compound selected from a group consisting of

organic carboxylic acids and amines.

34. The process according to claim 33, wherein said complexing agent (c) comprises at least one kind of a compound selected from a group consisting of citric acids, ethylenediaminetetraacetic acid, and salts thereof.

35. The process according to any of claims 1 to 27, wherein said reducing agent (e) comprises a material having an electrode potential which is inferior by more than 0.1 V to that of said metal (a') or said transition metal (b') which is the lowest.

36. The process according to any of claims 1 to 27, wherein said reducing agent (e) comprises a material having an electrode potential which is inferior by more than 0.2 V to that of said metal (a') or said transition metal (b') which is the lowest.

37. The process according to claim 30, wherein said reducing agent (e) comprises a material having a property that an aqueous solution of said material maintained at 25 °C exhibits a normal electrode potential of less than -0.2 V.

38. The process according to claim 30, wherein said reducing agent (e) comprises a material having a property that an aqueous solution of said material maintained at 25 °C exhibits a normal electrode

potential of less than -0.5 V.

39. The process according to any of claims 35 to 39, wherein said reducing agent (e) comprises at least one kind of a compound selected from a group consisting of lower oxygen compounds such as hypophosphorous acid, phosphorous acid, sulfurous acid, thiosulfuric acid, and dithionous acid; salts of these compounds; metal salts in the low valence state of Fe (II), Ti (III) and Cr (II); organic compounds such as formaldehyde, formic acid, and oxalic acid; and salts of these organic compounds.

40. The process according to any of claims 1 to 27, wherein said reducing agent (e) is soluble in said solvent (d).

41. The process according to any of claims 1 to 27, wherein said solvent (d) comprises at least one kind of a solvent selected from a group consisting of water and polar solvents.

42. The process according to claims 41, wherein said polar solvent includes alcohols, ester compounds, amide compounds, nitrile compounds, amine compounds, halogen compounds, sulfur compounds, liquid ammonia, and glacial acetic acid.

43. The process according to any of claims 1 to 27, wherein said solvent comprises at least one kind of a solvent selected from a group consisting of water and

alcohols.

44. The process according to any of claims 1 to 27, wherein said mixed solution obtained in said step (1) contains a complex formed by said metal (a') and said complexing agent (c).

45. The process according to any of claims 1 to 27, wherein said mixed solution obtained in said step (1) contains a complex formed by said transition metal (b') and said complexing agent (c).

46. The process according to any of claims 1 to 27, wherein the step (2) of mixing the reducing agent (e) with the mixed solution obtained in the step (1) is performed under condition with a temperature of -10 to 100 °C.

47. The process according to any of claims 1 to 27, wherein the step (2) of mixing the reducing agent (e) with the mixed solution obtained in the step (1) is performed under condition with a temperature of 10 to 90 °C.

48. The process according to any of claims 1 to 27, wherein the step (3) of oxidizing the reducing agent (e) contained in the mixture obtained in the step (2) is performed under condition with a temperature of -10 to 100 °C.

49. The process according to any of claims 1 to

27, wherein the step (3) of oxidizing the reducing agent (e) contained in the mixture obtained in the step (2) is performed under condition with a temperature of 10 to 90 °C.

5 50. The process according to any of claims 1 to 27, wherein the oxidation of the reducing agent (e) in the mixture in the step (3) is performed by adjusting said mixture to have a pH value in a range of from 3 to 12.

10 51. The process according to any of claims 1 to 27, wherein the oxidation of the reducing agent (e) in the mixture in the step (3) is performed by adjusting said mixture to have a pH value in a range of from 5 to 10.

15 52. The process according to any of claims 1 to 27, wherein the step (3) of oxidizing the reducing agent (e) contained in the mixture obtained in the step (2) is performed by adding an alkali to said mixture.

20 53. The process according to claim 52, wherein the addition of said alkali in the step (3) is performed so that the pH value of the mixture is changed from less than 2 to a value in a range of from 3 to 12.

25 54. The process according to claim 52, wherein the addition of said alkali in the step (3) is performed so that the pH value of the mixture is changed from less than 2 to a value in a range of from 5 to 10.

55. The process according to claim 52, wherein

the alkali comprises at least one kind of a compound selected from a group consisting of hydroxides of alkali metals, hydroxides of alkaline earth metals, amines, and ammonia.

5 56. The process according to any of claims 1 to 27, wherein the step (2) is performed in an atmosphere comprising at least one kind of a gas selected from a group consisting of hydrogen gas, nitrogen gas, and inert gas such as argon gas and helium gas.

10 57. The process according to any of claims 1 to 27, wherein the step (3) is performed in an atmosphere comprising at least one kind of a gas selected from a group consisting of hydrogen gas, nitrogen gas, and inert gas such as argon gas and helium gas.

15 58. The process according to any of claims 1 to 27, wherein the metal compound (a) and the transition metal compound (b) are used respectively in such an amount that [the number of moles of the metal (a') in the metal compound (a)]/[the number of moles of the transition metal
20 in the transition metal compound (b)] falls in a range of from 0.1 to 10.

25 59. The process according to any of claims 1 to 27, wherein the metal compound (a) and the transition metal compound (b) are used respectively in such an amount that [the number of moles of the metal (a') in the metal

compound (a)]/[the number of moles of the transition metal in the transition metal compound (b)] falls in a range of from 0.2 to 5.

5 60. The process according to any of claims 1 to 27, wherein the complexing agent (c) is used in such an amount that that [the number of moles of the complexing agent(c)]/[the number of moles of the metal compound (a) + the number of moles of the transition metal compound (b)] falls in a range of from 1 to 5.

10 61. The process according to any of claims 1 to 27, wherein the complexing agent (c) is used in such an amount that that [the number of moles of the complexing agent(c)]/[the number of moles of the metal compound (a) + the number of moles of the transition metal compound (b)]
15 falls in a range of from 2 to 5.

20 62. The process according to any of claims 1 to 27, wherein the reducing agent (e) is used in an amount which is 1 to 3 times versus the sum of the equivalence point of the metal compound (a) and that of the transition metal compound (b).

25 63. The process according to any of claims 1 to 27, wherein the reducing agent (e) is used in an amount which is 1 to 2 times versus the sum of the equivalence point of the metal compound (a) and that of the transition metal compound (b).

64. The process according to any of claims 1 to 27 which further includes a step of washing said amorphous alloy material.

65. The process according to any of claims 1 to 27 which further includes a step of drying said amorphous alloy material.

66. The process according to any of claims 1 to 27 which further includes a step of grinding said amorphous alloy material.

10 → 67. An electrode structural body comprising an electrode material produced by the process defined in any of claims 1 to 66 and a collector constituted by a material incapable of being alloyed with lithium in electrochemical reaction.

15 68. An electrode structural body according to claim 67, wherein a layer comprising said electrode material is formed on said collector.

20 69. An electrode structural body according to claim 67, wherein a layer comprising said electrode material and a binder is formed on said collector.

70. An electrode structural body according to claim 67, wherein a layer comprising said electrode material, an electrically conductive auxiliary, and a binder is formed on said collector.

25 71. An electrode structural body according to

claim 69 or 70, wherein the binder comprises a water-soluble organic polymer material.

72. A process for producing an electrode structural body, including a step of arranging an electrode material
5 produced by the process defined in any of claims 1 to 66 on a collector constituted by a material incapable of being alloyed with lithium in electrochemical reaction.

73. The process according to claim 72, wherein said electrode material is arranged on said collector by way of
10 press-forming.

74. The process according to claim 72 which comprises a step of preparing a paste by mixing said electrode material with a binder and, if necessary, a solvent and a step of arranging said paste on said
15 collector.

75. A rechargeable lithium battery comprising at least an anode, a cathode and an electrolyte and in which oxidation-reduction reaction of lithium is used, characterized in that said anode comprises an electrode
20 structural body defined in any of claims 67 to 71.

76. A rechargeable lithium battery according to claim 75, wherein said cathode comprises a lithium-containing electrode material.

77. A process for producing a rechargeable lithium
25 battery comprising at least an anode, a cathode and an

electrolyte and in which oxidation-reduction reaction of lithium is used, characterized by including a step (i) of forming said anode by arranging an electrode material produced by the process defined in any of claims 1 to 66 on a collector comprising a material incapable of being alloyed with lithium in electrochemical reaction to form an electrode structural body as said anode and a step (ii) of arranging said anode and said cathode to oppose to each other through said electrolyte.

78. The process according to claim 77, wherein the step (i) includes a step of arranging said electrode material on said collector by way of press-forming.

79. The process according to claim 77, wherein the step (i) includes a step of preparing a paste by mixing said electrode material with a binder and, if necessary, a solvent and a step of arranging said paste on said collector.

80. The process according to claim 77 which further includes a step of forming said cathode using a lithium-containing electrode material.